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Looking at pore scale processes in geomaterials using time-resolved 3D imaging and multi-scale imaging

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ABSTRACT

The study of transport and degradation processes in porous geological materials bears importance to a variety of real-world problems, both in underground as above ground. To fully comprehend the impact these processes have on large geological entities, it is crucial to understand what happens on the scale where fluids are transported through the sample and interact with the minerals in the geological material. New insights gained on the pore scale level allow for a better comprehension of processes and effects on larger scale and can be used as input for larger scale models.

Over the past decade, the availability of high quality laboratory-based X-ray micro-computed tomography (micro-CT) scanners has enabled many researchers to image and analyze the pore space of geological materials in 3D. However, a number of important challenges in both the acquisition and the analysis of 3D pore space information persists. On the one hand, specialized imaging, analysis and modeling techniques are needed to deal with the multi-scale aspect of many geo-materials. On the other hand, understanding the dynamics of pore-scale processes requires in-situ, time-resolved imaging. We will present the progress on these two key issues at Ghent University's Centre for X-Ray Tomography (UGCT). As an approach to tackle the multi-scale problem, we will show results from new complementary high-resolution 3D imaging techniques (e.g. ptychographic tomography). With regard to the challenge of time-resolved imaging, we will demonstrate our advances in (fast) in-situ, lab-based micro-CT imaging of transport and degradation processes in porous geo-materials (e.g. two-phase flow, salt crystallization, frost action, reactive flow).

The pore space in geo-materials is often very complex and spans multiple orders of magnitude going from larger centimeter scale pores (e.g. vuggy porosity) to nanometer scale pores in inside minerals (e.g. clays and micrite). Micro-CT can cover the centimeter to micrometer scale but lacks the ability to analyze features below the micrometer. Combining micro-CT with other 3D techniques like focused ion beam scanning electron microscopy (FIB-SEM) or ptychography are needed

to analyze the entire range of pores in a geo-material. Ptychographic tomography enables non-destructive 3D imaging at resolutions that were only achieved by FIB/SEM in the past. The method was used to study the structure and composition of clay mineral samples at resolutions down to 45 nm. Furthermore, the sample's response to changing relative humidity conditions could be monitored. As in most high-resolution techniques, sample size is very limited, in this case to 25 – 50 µm. We have shown the expansion of clay minerals from low to high relative humidity, and were able to exactly determine the different minerals present in the analyzed samples, together with their spatial distribution (Figure 1).

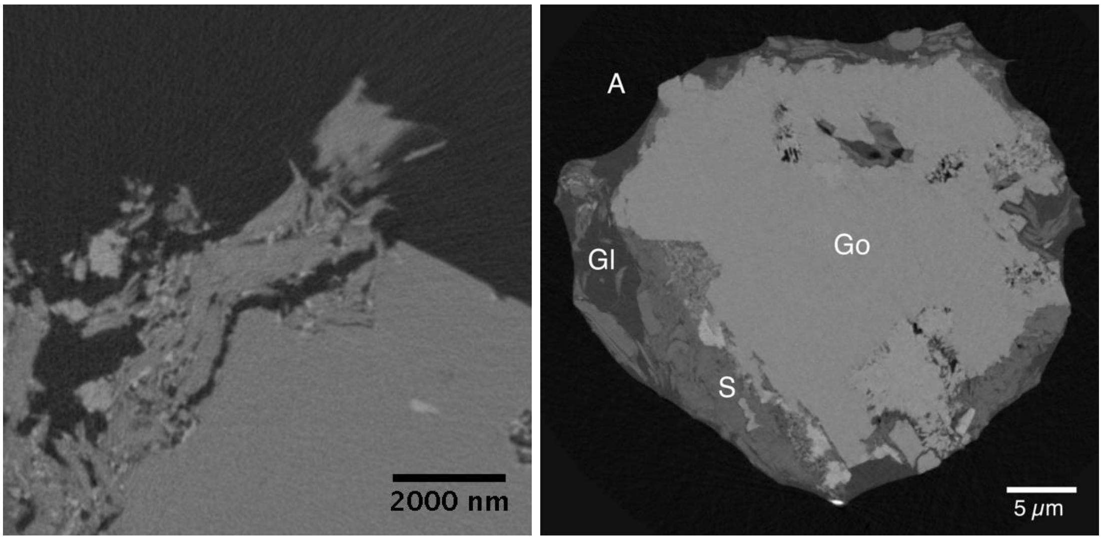

Figure 1: Left: Detail from ultra-high resolution(45 nm) reconstruction of Vosges sandstone. Right: Identification of different mineral phases; S = Smectite, Go = Goethite, Gl = sample mounting glue, A = air.

Apart from the constant improvement with respect to resolution and image quality, the temporal resolution of micro-CT imaging is also progressing, which results in shorter measurement times (down to 10 seconds for a full 360° acquisition). The resulting shorter measurement times allow to perform dynamic, high resolution 3D visualization of the sample and basically implies that instead of a 3D image of the internal structure of the sample, a 4D video at high spatial resolution can be obtained. This development therefore permits to monitor changes and alterations in a geo-material in real time when this material is exposed to certain environmental conditions like pressure, temperature and relative humidity. In-situ scanning or scanning the geo-material while it is exposed to external conditions requires specific add-on modules. At the UGCT multiple add-on modules like high pressure cells (up to 100 bar), uniaxial compression stages (up to 5kN), triaxial flow cells, relative humidity chambers and freezing stages were developed in the past two years to allow in-situ imaging at high spatial resolutions (down to 5µm). A few geological applications of in-situ dynamic micro-CT imaging are discussed below.

Application 1: Diffusive and advective transport of a tracer salt in an oolithic limestone.

The solute transport and preferential flow inside the pore space of the Savonnières limestone was visualized by pumping a highly X-ray attenuating brine (containing 10 wt.% CsCl) into a water-saturated sample at a flow rate of 0.6 ml/min. The sample had a diameter of 6 mm and was placed in a custom built PMMA flow cell. The limestone has a dual porosity with well-connected pores between the grains (intergranular porosity) and secondary porosity inside the grains (intragranular porosity), which is connected to the rest of the pore network through microporosity.

Prior to the dynamic flow experiment the limestone sample was fully saturated with water and a high-quality micro-CT scan was performed (voxel size 7.4 µm) to obtain a better characterization of the pore network in the sample. Afterwards, while an X-ray attenuating brine was pumped into to the sample, a fast micro-CT scan was acquired every 12 seconds for a total period of 5 minutes. The fast scans were recorded and processed with the proprietary 4D tools of the ACQUILA software (XRE, Ghent, Belgium) which resulted in a total of 72 reconstructions at a voxel size of 14.8 µm.

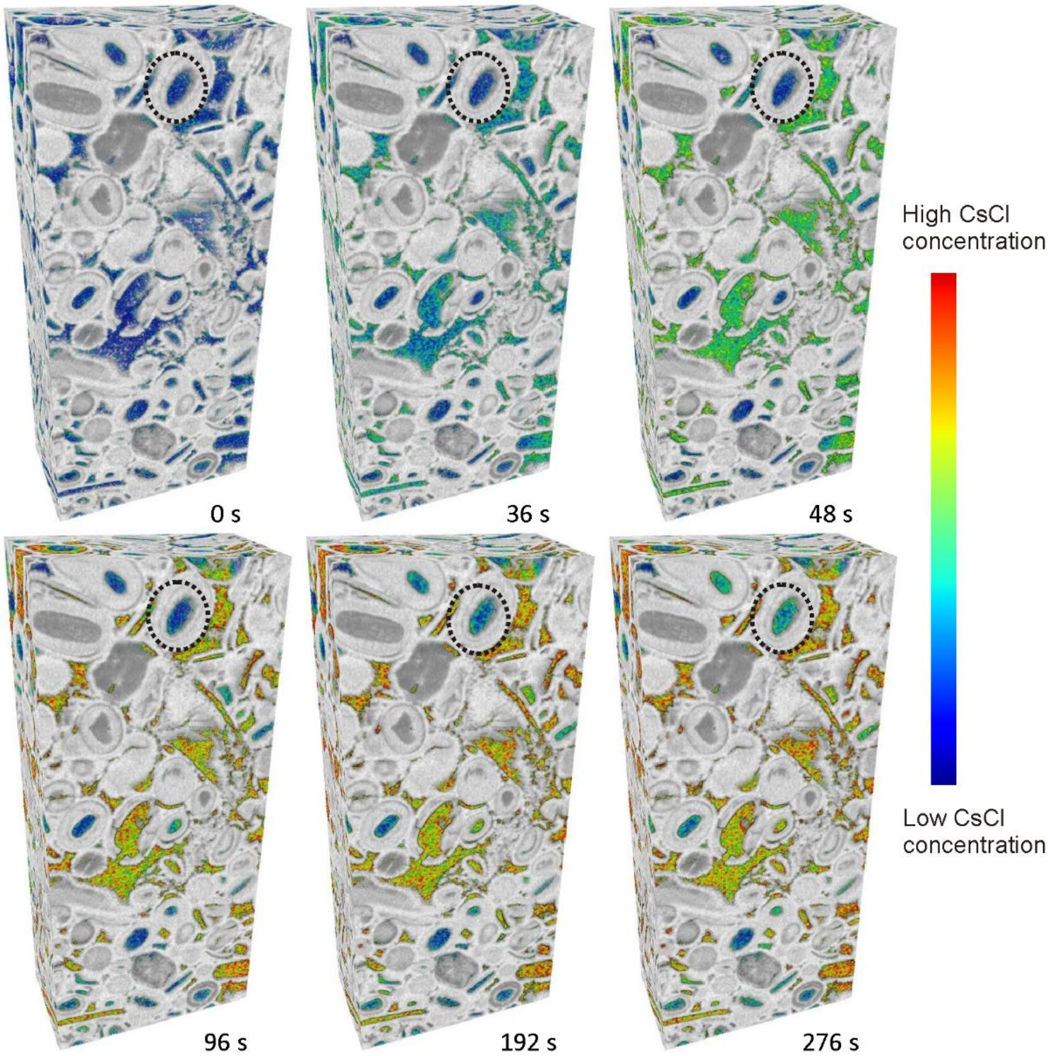

Figure 2. The renderings illustrate the evolution of the CsCl concentration in the macropores. The dotted circle indicates an intragranular pore in which the flow is stagnant. Time series measurements were performed with the EMCT-scanner at an acquisition time of 12 seconds.

In figure 2, renderings of the limestone and the pore space are shown at different time steps during brine injection. The pore space is colour coded according to the CsCl concentration. At 0 seconds no CsCl has entered the pore space, which is illustrated by the homogeneous blue colour. After 96 seconds, the CsCl is heterogeneously distributed and most of the intergranular pore space contains a high CsCl concentration, while the intragranular pores in the dissolved ooids (figure 2, dotted circle) still have a low concentration. This illustrates that the distribution of the CsCl in the intergranular pore space is mainly controlled by advection and these pores are preferential flow paths in the limestone. Over time there is a more gradual increase of the CsCl concentration in the intragranular pores and the CsCl concentration becomes more homogeneous. This more stagnant behaviour indicates that solute transport in these pores is mainly controlled by diffusion and illustrates that important differences in fluid chemistry arise between the more stagnant regions (intragranular pores) and the preferential flow paths with a higher advective transport rate.

Application 2: Freeze-thaw cycling and fracture formation in limestone.
Freeze−thaw cycling stresses natural building materials and can be responsible for fracture formation in the porous geo-material. Understanding and predicting the effect of freeze−thaw cycles is therefore important in the built environment and cultural heritage preservation. In-situ dynamic micro-CT imaging, using a special designed freezing stage, is applied to study the pore scale dynamics related to freezing and thawing. The experiments show the development of a fracture network in a porous limestone. During freezing fractures are opened when cooling below −9.7 °C, which is the moment at which ice crystallizes in the fracture. During thawing the fractures close again and are less pronounced in the micro-CT image (figure 3). After multiple freezing and thawing cycles however the fractures reach a critical volume resulting in a residual strain. Laboratory, continuous X-ray micro-CT scanning opens new perspectives for the pore-scale study of ice crystallization in porous media as well as for environmental processes related to freeze−thaw fracturing.

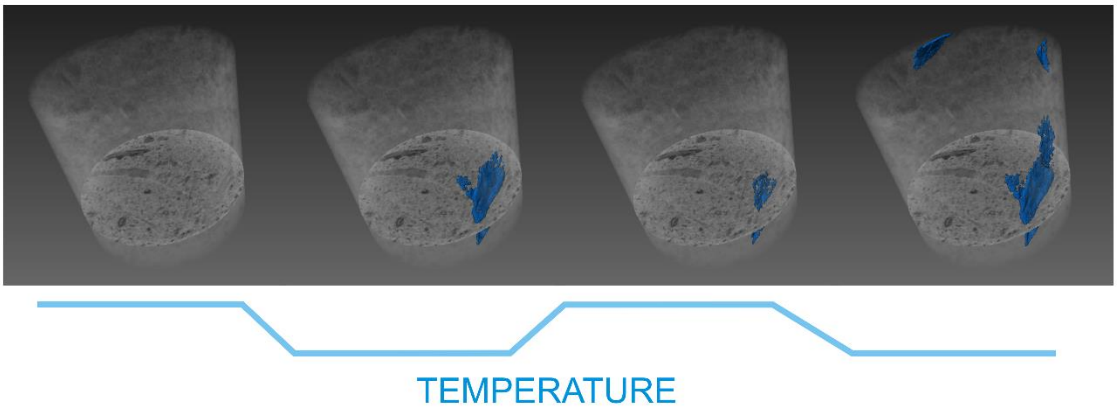

Figure 3. 3D rendering of limestone with a fracture in blue. The fracture opens during freezing and closes during thawing.

Application 3. Drying and salt growth in sandstone.
The drying dynamics of salt loaded Mšené sandstone were studied at 20% RH and 50% RH on samples of 8 mm in diameter and 10 mm in height. The samples were initially scanned in their dry state. Next, the samples were capillary saturated by immersing them in a saturated NaCl solution for 30 minutes. The samples were then scanned in their wet state, and subsequently during their drying, by scanning them every 30 minutes during the first 3 hours, and every hour during the

succeeding 12 hours. Drying was controlled by placing the sample in a custom-built climatic chamber, compatible with the µCT setup. Each scan took 9 minutes, and the experiments resulted in scan series with a voxel size of 10^3 µm³. The datasets were further analysed with the software Avizo (FEI).
Drying at 20% RH is initially faster than at 50% RH, due to the higher difference in RH between the sample's surface and the environment (figure 4a). This first drying stage is characterized by a constant rate, due to the capillary action which induces a hydraulic connection between the surface and the inner pore space. Subsequently, the evaporation at the surface mainly results in salt efflorescence. At 50% RH, this constant rate period lasts until the sample is almost completely dried out. At 20% RH, the drying changes to a slower, exponential regime after about 1 hour, i.e. after approximately half of the initial salt solution has dried out.

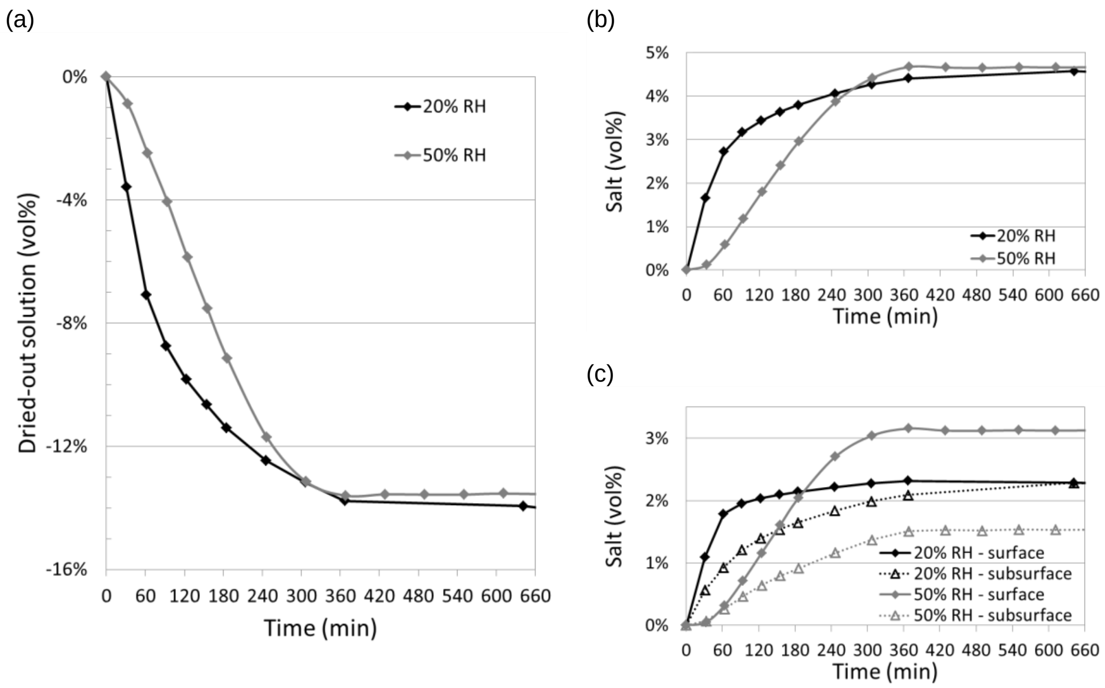

Figure 4. The drying (a) and crystallization dynamics (b-c) of Mšené sandstone at 20% RH and 50% RH. Fig 2c disseminates between the salt crystals forming on the outer surface (efflorescence) and the salt crystals precipitating just below, in the pores of the subsurface.

The differences in drying kinetics are caused by the differences in salt precipitation dynamics, which could be, for the first time, characterized simultaneously in 4D using dynamic µCT. While drying at 20% RH, the crystal growth on the surface (efflorescence) develops much faster than at 50% RH (figures 4b-c), mainly during the first hour of the experiment, and then reaches a plateau. The salt crystals precipitating just below the surface at 20% RH develop more gradually, during the whole duration of the drying. The period of the formation of efflorescence at 20% RH is congruent with the period of the constant drying rate in the drying curve. The second exponential drying stage at 20% RH must thus be resulting from this salt efflorescence, corroborating the statement that a salt skin is forming on the surface during the first stage of drying (Desarnaud et al. 2015), partially closing the pores and causing a slower drying during the continuation of the process. During this second period, most of the salt crystals precipitate just below this skin layer. Typical snapshots of the sample drying at 20% RH are given in figure 5. At 50% RH, the crystal precipitation develops slower, and does not block the drying.

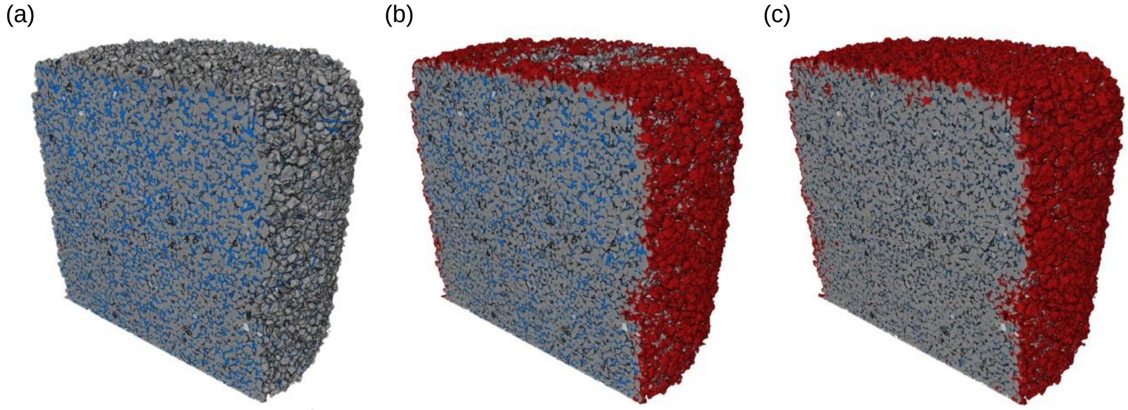

Figure 5. Typical snapshots of the Mšené sandstone sample while drying at 20% RH at the wet state (a), after 1 hour (b) and after 6 hours (c). Blue: salt solution – red: salt crystals.

Acknowledgements

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